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INDIANA UNIV AT BLOOMINGTON DEPT OF PHYSICS
OPTICAL PUMPING STUDIES OF COLLISIONAL INTERACTIONS. (U)

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Optical Pumping Studies of Collisional Interactions

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I. Resume of Results of Research

The primary goal in our AFOSR sponsored research has been the extension of previously developed techniques of optical pumping to the study of certain kinds of atomic collisional interactions. We have been particularly interested in studying various modes of relaxation of electronic and nuclear spin polarizations induced in collisions of magnetically polarized atoms with foreign (buffer gas) atoms. Such studies have practical significance in providing insight into the physical phenomena which govern the operation and performance of magnetometers, masers, nuclear gyroscopes, and secondary frequency standards. They are of fundamental interest through the unique information they provide on interatomic interactions.¹

We have solved many problems enroute to fulfilling the goals set forth in our original proposal to AFOSR. Of particular significance is our illucidation of the previously unsuspected dominant role played by quasi molecular collisions in the relaxation of alkali metal atoms in the light noble gases. In the following narrative I summarize our progress and contributions in the several areas in which we have worked. Full details and references to the work of others can be found in our publications listed in later sections.

A. Optical Pumping Transients in Weak Magnetic Fields

1. Background

Electronic and nuclear spin polarizations are produced in optical pumping through the absorption and re-emission of circularly polarized resonance radiation. These polarizations correspond to artificially created imbalances in the relative populations of the Zeeman sublevels of

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atomic ground states and as such find practical application in atomic frequency standards, magnetometers, gyroscopes, and masers. The degree of polarization attained and the exact distribution of relative populations produced throughout the Zeeman sublevels depends upon the pumping process itself and upon a variety of forms of collisional relaxation, both in the ground and excited atomic states. In all such processes the hyperfine interaction between the electronic magnetic moment and the nuclear spin can play an extremely important and complicating role.²⁻⁷

One can observe the evolution of a system undergoing optical pumping by monitoring the intensity of light scattered or transmitted by the optical pumping cell. Under the appropriate conditions the time constants of the transient pumping signals directly reflect the characteristics of the various relaxation processes. In the course of our AFOSR sponsored research we brought together all present relevant knowledge of the mechanics of the pumping and relaxation processes and obtained analytic solutions describing pumping transients in alkali metal vapors.⁸ We showed rigorously within well established approximations how collisions of pumped atoms with the walls of the experimental cell, simple binary alkali-atom-buffer-gas atom collisions, and alkali-alkali spin exchange collisions all contribute to determining the shape of these transients. We included rigorous treatments of the effect of the hyperfine interaction on all pumping and collisional relaxation processes. We showed that in low magnetic fields the pumping transients of the electronic spin polarization are generally represented by the sum of two exponentials, the rate constants of which depend wholly upon relaxation processes in the ground state, with the relative contributions of the two rates to the production of the ground state electronic spin

polarization ($\langle S_z \rangle_g$), being determined mainly by the degree of collisional relaxation which occurs during the excited state lifetime. The comprehensive analytic expressions we derived involved all known relaxation processes: they offered new and dependable ways to measure cross sections for electronic and nuclear spin relaxation in sudden binary collisions between alkali metal atoms and noble gas atoms both in the ground and excited states of the alkali metal atoms.

The calculations described above were made subject to the assumptions that equal intensities are present in all absorbable components of the pumping line, and that the pumping rate is small compared to relaxation rates. These are requirements that often were postulated or assumed in many optical pumping experiments performed in the past, but which virtually never were satisfied in practice. The principal problem is that the light emitted by conventional resonance lamps is very seldom "flat" and of equal intensity over all hyperfine components of the pumping line. Even if an appropriate match of the line shape of the pumping radiation and the line shape of this absorption cell can be made at a particular buffer gas pressure, this match is destroyed at higher buffer pressures where the absorption line of the optical pumping cell is considerably broadened. We therefore put considerable effort into extending a new optical pumping technique utilizing filtered white light as the pumping source. The experimental conditions in this technique satisfy exactly the requirements of our weak pumping limit calculations. We first obtained extensive experimental results for Cs which provided well defined values of the cross sections for collisional relaxation both in the ground and excited states and of

the Cs-buffer gas atom mutual diffusion coefficients. The experimental results verified our theoretical calculations, and the combination of theory and experiment cleared up many previously unresolved questions remaining from earlier experiments.⁸ There remained one puzzling discrepancy, however, a factor two difference between our measurement of the Cs-Ne nuclear spin independent cross section for $\langle S_z \rangle$ relaxation and an equally reliable measurement of the cross section for $\langle S \cdot I \rangle$ relaxation made independently by an Italian group. Our desire to resolve this discrepancy, and to test further the validity of our theoretical calculations and experimental techniques led us to extend our work to the measurements and analyses of pumping transients in ^{85}Rb and ^{87}Rb described below.

2. The effect of the formation and destruction of quasi-bound Rb-He and Rb-Ne molecules on electronic and nuclear spin relaxation in Rb^{85} and Rb^{87}

In 1976 we reported on the extension of our white light optical pumping experiments to the measurement of relaxation parameters in Rb^{85} and Rb^{87} .⁹ An important initial motivation was to capitalize upon the expertise we had developed in order to secure definitive values both of the nuclear spin independent cross sections (σ) for the relaxation of $\langle S_z \rangle$ of Rb in sudden collisions of Rb atoms with buffer gas atoms, and of the diffusion coefficients (D_0) of Rb atoms in the various buffer gases. Toward this end we measured and analyzed over a thousand optical pumping transients in He, Ne, Ar, and N_2 buffer gases at pressures ranging from 0.5 Torr to over 600 Torr. We obtained the sought for values

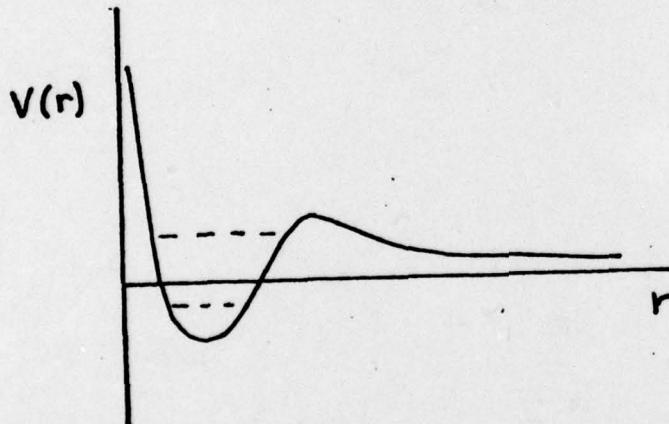
of D_0 and σ , but in doing so discovered an effect which significantly alters the previous understanding of the collisional interaction responsible for spin relaxation in Rb.

Taking all "normal" relaxation processes into account, theory predicted that the measured differences between the two rate constants, $(Z_2 - Z_1)$ of the $\langle S_z \rangle$ pumping transient should depend only upon σ and the spin exchange cross section σ_s , together with various well known or easily calculable parameters. Our measurements in Rb-He and Rb-Ne, however, indicated the presence of an anomalous effect: there was a large additional contribution to relaxation, R^* , which could not be attributed to any normal relaxation mechanism. Indeed our experimental results for Rb in He indicated that at the commonly used buffer gas pressures of 10 Torr to 100 Torr, the anomalous relaxation rate is more than an order of magnitude greater than that arising from normal binary collisions! We have since been able to show that this anomalous relaxation almost certainly is due to the formation of quasi-bound Rb-He and Rb-Ne molecular complexes.¹⁰ In the paragraphs below I summarize the significance and importance of this work.

It was assumed in the past that spin relaxation in sudden collisions of alkali atoms with light noble gas atoms occurs through a spin-orbit interaction ($\gamma(\vec{S} \cdot \vec{N})$) generated in sudden alkali-atom-buffer-gas atom collisions. Such sudden collisions last a time of the order of that required for the two atoms to "fly by" each other, in other words, about 10^{-13} sec at 300°K. Other kinds of collisions also are possible, however. If, for example, an alkali atom collides simultaneously with two noble gas atoms, a weakly bound alkali-atom-noble-gas-atom van der Waals molecule may be

formed with the third atom carrying off the appropriate amount of excess energy.¹¹ Such a molecule generally will live only until hit by another buffer gas atom. At pressures of the order of several Torr this time may be of the order of 10^{-7} sec, very long compared to the duration of a fly-by collision. The significantly longer duration of perturbation in the molecular case can result in much greater probability of relaxation. Thus even though events of molecular formation may be relatively rare, they may still have great influence ultimately on spin relaxation rates.

Still a third type of alkali-noble-gas collision is possible. The effective potential for an alkali-atom-noble-gas-atom pair is the combination of the van der Waals potential and the centrifugal barrier potential: the sum is shown schematically below.



True bound states formed in three body collisions are those with energies less than zero on the $V(r)$ scale. There are the possibilities of quasi-bound states, however, with energies greater than zero, but less than the height of the barrier potential. Such quasi-bound molecules can be formed in the same way that normal bound molecules are formed, i.e. in three body

collisions. They also can be formed in two body collisions, i.e. an alkali-atom-buffer-gas-atom pair of the appropriate total energy can "tunnel" through the barrier to form a quasi-bound state. Quasi-bound complexes should behave much as the bound states except for the fact that they have a natural dissociation (tunneling) rate in addition to the collisional destruction rate.

Contributions of molecular formation to the rate of spin relaxation of Rb in the heavy noble gases Kr and Xe had already been demonstrated and studied extensively by Bouchiat and co-workers.¹¹ They showed that the relaxation mechanism was the same as that present in binary collisions, namely $\gamma(\vec{S} \cdot \vec{N})$, acting for a much longer time. It was evident, however, that the anomalous effects we saw in Rb-He and Rb-Ne must arise from a different source. Predictions based on the $\gamma(\vec{S} \cdot \vec{N})$ interaction suggest that relaxation rates should become progressively larger as one moves from light to heavier noble gases. Since rates of molecular formation also are larger in the heavier noble gases, one predicts a far greater rate of $\gamma(\vec{S} \cdot \vec{N})$ relaxation for alkali metals in the heavier noble gases than in the lighter gases. Indeed, the rate for Rb in Ar should be of the order of 10^4 times greater than that for Rb in He. Our results, however, indicated essentially equal rates of anomalous relaxation for Rb in He, Ne, and Ar. The nuclear spin dynamics associated with anomalous relaxation also were incompatible with $\gamma(\vec{S} \cdot \vec{N})$ relaxation. In the case of $\gamma(\vec{S} \cdot \vec{N})$ relaxation via molecular formation, theory predicts strong contributions of the anomalous relaxation rate to both exponential terms in the $\langle S_z \rangle_g$ relaxation transient. In practice, however, we found a strong contribution to the "fast" rate constant, but an essentially zero contribution to the

"slow" rate constant. There is no way to force the $\gamma(\vec{S} \cdot \vec{N})$ interaction in molecular formation to fit the observed experimental facts.

We were able to show, however, that another interaction present both in binary collisions and in molecular collisions, the modification of the hyperfine interaction of the alkali atom due to the proximity of a buffer gas atom, $\delta a(\vec{S} \cdot \vec{I})$, indeed provides an explanation of the source of anomalous relaxation. For example, in contrast to the case of the $\gamma(\vec{S} \cdot \vec{N})$ interaction, the $\delta a(\vec{S} \cdot \vec{I})$ interaction is strongest in the He and weakest in Ar. Taking other factors such as the formation rates of molecular complexes into account, one predicts essentially equal anomalous relaxation rates in Rb-He, Rb-Ne, and Rb-Ar, in accord with the experimental observations. Much more dramatic, however, are the calculations of Z_1 , the "slow" rate constant in the $\langle S_z \rangle_g$ transient, and the relative weights, D_1/D_2 , of the coefficients of the two exponential terms of the transient. Excellent agreement is attained throughout a wide range of buffer gas pressures when the peculiar nucleon spin dynamics associated with the $\delta a(\vec{S} \cdot \vec{I})$ interactions are introduced. The essentially zero contribution of anomalous relaxation to Z_1 , for example, is fully explained.¹⁰

The identification of anomalous relaxation in Rb-He and Rb-Ne, and consequently in Cs-Ne, allowed us to resolve ambiguities and uncertainties in reported cross sections which amounted to differences greater than two orders of magnitude. A comprehensive review of our work and the rationalization of earlier measurements is contained in Refs. 9 and 10.

3. Measurement of electronic and nuclear spin relaxation in the $^2S_{1/2}$ ground states of K³⁹ and K⁴¹.

Following our discovery of anomalous relaxation in Rb, we began

a study of spin relaxation in optically pumped potassium vapor. Part of this work was completed during the term of the grant award; part has continued into the present year. The motivation for the experiment follows.

An approximate treatment of anomalous relaxation via the $\delta a(\vec{S} \cdot \vec{I})$ interaction in van der Waals molecular formation yields Eq. 1:⁹

$$R^*(\delta a(\vec{S} \cdot \vec{I})) \approx \frac{(\text{Const}) \tau_{e2}^2 T_f^{-1} \left(\frac{\omega_s}{\Delta W} \right)}{1 + (\Delta W)^2 \tau_{e2}^2} , \quad (1)$$

where $(\tau_{e2})^{-1}$ is the rate of breakup of molecular complexes, T_f^{-1} is their formation rate, ΔW is the angular hyperfine frequency, and ω_s is the electronic Larmor frequency in the external magnetic field (H_0). The availability of two isotopes of potassium, K^{39} and K^{41} , both with hyperfine frequencies more than an order of magnitude less than those of the Rb isotopes, makes possible the study of the predicted dependence of R^* upon ΔW . It also is important to determine the dependence of the R^* term upon buffer gas pressure. At high gas pressures, that is, at high breakup rates, molecular collisions must approach the limit of sudden binary collisions, and the $(\omega_s/\Delta W)^2$ factor must decrease, as must R^* . We calculated that K should show this effect more readily than Rb. Our experimental results suggest that it may be possible in K-Ne to separate the effects of the formation of bound and quasi bound molecular interactions. Further measurements are in progress. Additional motivation for this work is that the only previous measurement of K relaxation was made before even normal nuclear spin effects were fully understood. The accuracy of the reported cross section therefore is questionable.

Work actually completed to date, much of it during the term of the grant, includes the following: Measurement of double exponential relaxation of K³⁹ and K⁴¹ in He and Ne, and measurement of K³⁹ and K⁴¹ single exponential relaxation in low pressures of He and Ne. Diffusion coefficients and nuclear spin independent cross sections for the relaxation of $\langle S_z g \rangle$ have been evaluated. Anomalous rates of relaxation have been extracted. Considerable efforts to obtain measurements on the rare isotope, K⁴⁰, were not successful. That is particularly unfortunate since the anomalously large nuclear spin (I = 4) and hyperfine structure of this isotope promised useful comparisons with measurements on the more prevalent isotopes.

B. Optical pumping in high magnetic fields; contributions of the $\delta a(\vec{S} \cdot \vec{I})$ collisional interaction to relaxation rates of Cs in He and Ne.

In earlier AFOSR sponsored research we performed an experiment involving the optical pumping of cesium in magnetic fields ranging up to 100kG. This work constituted the first study of collisional relaxation of electronic spin polarization in very high magnetic fields. The experiment was completed and published in 1977, and provided some intriguing discoveries. First, in strong magnetic fields (of the order of 20kG or greater), one should be able to measure electron spin relaxation fully decoupled from effects of nuclear spin dynamics. We verified this in Cs-Ar, where we measured a relaxation cross section of $110 \times 10^{-23} \text{ \AA}^2$. The high field value agrees well with our low magnetic field measurement of the nuclear spin independent cross section ($108 \times 10^{-23} \text{ \AA}^2$) and the measurement of $104 \times 10^{-23} \text{ \AA}^2$ for the cross section for relaxation of $\langle \vec{S} \cdot \vec{I} \rangle$ measured by Beverini et al. According to theory all these cross sections should be equal--they are, in fact, theoretically the same entity.

The results in Cs-Ne and Cs-He, however, were puzzling. Rather than finding a cross section equal to the nuclear spin independent cross section, we found equivalent high field cross sections to be approximately 2.5 times larger than the low field values. The full explanation of this effect is given in Ref. 12. Summarizing briefly, if the collisional spin orbit interaction, $\gamma(\vec{S} \cdot \vec{N})$, were the only interaction present between alkali metal atoms and buffer gas atoms, one should measure equal relaxation cross sections at low and at moderately high magnetic field. However, collisions also perturb the alkali metal atom hyperfine interaction, $\delta a(\vec{S} \cdot \vec{I})$, giving rise to the well known pressure shift of hyperfine frequencies in optically pumped masers and frequency standards. The $\delta a(\vec{S} \cdot \vec{I})$ interaction also contributes to relaxation rates at low or intermediate magnetic fields according to the following equation:

$$R_2(\delta a(\vec{S} \cdot \vec{I})) \approx \frac{8\pi^2}{3} (\delta v)^2 \frac{(n_o \sigma_c v_{rel}/p_o)^P}{(1 + \omega_o^2 \tau_{e2}^2)} \left(\frac{\omega_s}{\Delta W} \right)^2, \quad (2)$$

where δv is the measured hyperfine frequency shift in Hz/Torr, σ_c is an average cross section for the $\delta a(\vec{S} \cdot \vec{I})$ interaction in binary collisions, p is the buffer gas pressure in Torr, and ω_o , τ_{e2} , ω_s , and ΔW have been defined in section A above. At low magnetic fields the factor $(\omega_s/\Delta W)^2$ is small, generally making R_2 negligible compared to other relaxation rates. At fields above a few kG, however, the correct expression for R_2 is obtained by letting $(\omega_s/\Delta W) = 1$, that is,

$$R_2(\delta a(\vec{S} \cdot \vec{I})) \text{ (High } H_o) \approx \frac{8\pi^2}{3} (\delta v)^2 \frac{(n_o \sigma_c v_{rel}/p)^P}{1 + \omega_o^2 \tau_{e2}^2}. \quad (3)$$

Calculations show that for Cs-Ne and Cs-He the contributions of the $\delta a(\vec{S} \cdot \vec{I})$ interaction to relaxation rates at high magnetic field in fact are greater than those for $\gamma(\vec{S} \cdot \vec{N})$. The apparent "discrepancy", then, was caused by the contribution of a previously unsuspected relaxation process significant only at high magnetic field. Through analysis of our high field measurements we were able to make estimates of the average cross sections for the $\delta a(\vec{S} \cdot \vec{I})$ interactions in Cs-He and Cs-Ne, along with the average durations (correlation times) for these interactions in sudden binary collisions.

The results described above have led to extensions of high field experiments which were begun under the duration of the grant, but which are continuing at the present time. In particular, the following are of interest:

1. Measurement of the relaxation of $\langle S_z \rangle$ in the $^2S_{1/2}$ ground states of Rb^{85} and Rb^{87} at high magnetic field.

The only difference in Eq. 3 for Rb^{85} and Rb^{87} at high magnetic field lies in the factor (δv) : (δv) for Rb^{87} is approximately 2.25 times larger than that for Rb^{85} . Measurement of the relaxation rates of the separated isotopes at high magnetic field therefore is important for the confirmation of the $\delta a(\vec{S} \cdot \vec{I})$ interaction's contributions to relaxation rates.

Other kinds of information also can be acquired:

2. Measurement of the correlation time, τ_{e2} for the $\delta a(\vec{S} \cdot \vec{I})$ interaction.

With our magnetic field capability extending now to 140kG we hope to measure relaxation rates in the region where the term $\omega_0^2 \tau_{e2}^2$ in Eq. 3 is non negligible. If a fall-off of relaxation indeed can be seen at high magnetic field, that would lead to the first definitive measurement of τ_{e2} . This experiment would be performed in He or Ne, where the $\delta a(\vec{S} \cdot \vec{I})$ interaction is dominant.

3. Measurement of the correlation time, τ_{el} , for the $\gamma(\vec{S} \cdot \vec{N})$ interaction.

We plan measurements similar to those in a, above, but in Ar where the $\gamma(\vec{S} \cdot \vec{N})$ interaction should be dominant, possibly leading to a determination of τ_{el} for Rb-Ar.

4. Measurement of the average cross section for the $\delta a(\vec{S} \cdot \vec{I})$ interaction,

$$\sigma_c$$

At fields of the order of 40kG, $\omega_0^2 \tau_{el}^2 \ll 1$, and Eq. 3 reduces to

$$R_2(\delta a(\vec{S} \cdot \vec{I})) \approx \frac{8\pi^2}{3} (\delta v)^2 (n_0 \sigma_c v_{rel} / p_0) P. \quad (4)$$

Measurements of relaxation rates under these circumstances lead to values for σ_c . Again, we plan measurements on Rb in He and Ne, where the $\delta a(\vec{S} \cdot \vec{I})$ interaction should dominate at high magnetic field.

C. Relative probabilities for intermultiplet transitions.

In recent years there has been widespread interest in the study of collisionally induced transitions between atomic multiplet states, especially between the various $^2P_{1/2}$ and $^2P_{3/2}$ states of alkali metal atoms. It is of particular interest to measure experimentally the relative probabilities connecting particular Zeeman sublevels in such transitions, since such measurements provide tests of the various theoretical descriptions of the interaction responsible for collisional transfer. Some of our earlier AFOSR sponsored research, and a part of that reported in section A above, is connected with measurement of various cross sections for relaxation within a multiplet, i.e. within the $^2P_{1/2}$ state or $^2P_{3/2}$ state.^(6,8) The only way that full information on intermultiplet transition probabilities can be obtained, however, is to make a selective excitation of atoms in a

particular Zeeman sublevel, and observe the intensities of resonance fluorescence from other sublevels populated by collisional transfer. Experiments of this kind require both energetic resolution of the individual Zeeman components of the optical transition connecting the multiplet state with the ground state and decoupling of nuclear spin effects. High magnetic fields, of the order of 10kG or more therefore are required. In the presence of a magnetic field of the order of 100kG one also has the hope of splitting the various Zeeman components so far that the collisional interaction no longer possesses frequency components capable of inducing relaxation, thereby permitting measurement of the correlation time for the interaction.

During the grant period we completed a series of measurements between and among the Zeeman sublevels of the $^2P_{1/2}$ and $^2P_{3/2}$ transition probabilities of K in He and Ne. The experiment involved selective excitation of particular sublevels by narrow band dye laser radiation over magnetic fields ranging from 10kG to 30kG. These measurements yield cross sections for all collisionally induced transitions $|J, m_J\rangle \rightarrow |J', m'_J\rangle$, within the 4P multiplet for K colliding with He and Ne and are contained in a Ph.D. thesis by R. Boggy. Publication has been delayed, however, awaiting extension of the measurements to magnetic fields ranging up to 137kG. The latter measurements were made subsequent to the grant period and presently are being analyzed. Submission of the entire work for publication is anticipated within the present year.

1. For extensive reviews of many theoretical and experimental aspects of optical pumping relevant to the present work, see W. Happer, *Rev. Mod. Phys.* 44, 169 (1972), and "Optical Pumping", by L.C. Balling, in *Advances in Quantum Electronics* 3, ed. by D.W. Goodwin, Academic Press, New York (1975).
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8. F.A. Franz and C.E. Sooriemoorthi, *Phys. Rev. A* 8, 2390 (1973); A10, 126 (1974).
9. F.A. Franz and C. Volk, *Phys. Rev. Letters* 35, 1704 (1975); *Phys. Rev. A* 14, 1711 (1976).
10. F.A. Franz and C. Volk, *Phys. Rev. A* 18, 599 (1978).
11. See, for example, M.A. Bouchiat *et al.*, *J. Chem. Phys.* 56, 3703 (1972).
12. T.R. Marshall, F.A. Franz, and R. Boggy, *Phys. Rev. A* 16, 618 (1977).

II. Journal Articles Published During the Grant Period Reporting AFOSR**Sponsored Research**

1. "Spin Relaxation within the $6^2P_{1/2}$ and $6^2S_{1/2}$ States of Cesium

Measured by White Light Optical Pumping," F.A. Franz and C.E. Sooriamoorthi, *Physical Review A* 10, 126 (1974).

White light optical pumping techniques were applied to Cs-noble gas systems. Solutions to weak pumping rate equations were extended to include effects due to spin exchange. Collisional relaxation cross sections were measured in the $^2P_{1/2}$ and $^2S_{1/2}$ states of Cs. Cs-noble-gas diffusion coefficients were measured. Contributions to relaxation rates from possible formation of Cs-noble-gas molecules were studied.

2. "Relative Transition Probabilities in De-Excitation of Atomic

States by Collisional Quenching: $Cs^2P_{1/2} \rightarrow ^2S_{1/2}$," F.A. Franz, R. Boggy, and C.E. Sooriamoorthi, *Phys. Rev. A* 11, 1 (1975).

The effect of the hyperfine interactions on relative probabilities for quenching induced transitions between the Zeeman sublevels of the $^2P_{1/2}$ and $^2S_{1/2}$ states of alkali metal atoms was calculated and detailed analyses of the pumping equations were made.

Experimental measurements were performed in the Cs-N₂ system.

Enhancements of spin polarization due to quenching induced modifications of atomic de-excitation probabilities were discovered.

3. "Detection and Lifetime Measurement of Rb-He Quasi-Bound Molecules",

F.A. Franz and C. Volk, *Phys. Rev. Letters* 35, 1704 (1975).

Quasi-bound Rb-He van der Waals molecules were observed through

their influence on the electron spin relaxation of optically pumped Rb⁸⁵ and Rb⁸⁷. The natural lifetime of the complex was measured.

4. "Spin relaxation of rubidium atoms in sudden and quasi molecular collisions with light-noble-gas atoms", F.A. Franz and C. Volk, Phys. Rev. A 14, 1711 (1976).

Cross sections for electronic spin relaxation in sudden binary collisions of Rb atoms with He, Ne, Ar, and N₂ atoms were measured. Anomalous relaxation rates attributed to the formation of Rb-He and Rb-Ne van der Waals molecules were isolated and analyzed. Recognition of the anomalous effects was shown to resolve many disagreements in previously published studies of relaxation phenomena.

5. "Spin relaxation of optically pumped cesium in high magnetic fields", T.R. Marshall, R. Boggy, and F.A. Franz, Phys. Rev. A 16, 618 (1977).

The relaxation of optically pumped Cs was measured in magnetic fields ranging up to 100kG. The rate of electronic spin relaxation measured in Cs-Ar is approximately the same as that predicted by the low-field nuclear-spin-independent cross section for the relaxation of $\langle S_z \rangle$ vis the $\gamma(S \cdot \vec{N})$ (spin-orbit) interaction.

In the cases of Cs-He and Cs-Ne, considerably higher relaxation rates were measured at high fields than those that are predicted from the low-field cross sections. The differences were shown to be due to relaxation arising from the collision-induced modi-

fication of the Cs hyperfine interaction, $\delta a(\vec{S} \cdot \vec{I})$. Upper limits of the order of 10^{-13} sec were set for the average durations of both the $\delta a(\vec{S} \cdot \vec{I})$ and the $\delta(\vec{S} \cdot \vec{N})$ interactions in sudden binary collisions of Cs atoms with light-noble-gas atoms. The average cross sections for the $\delta a(\vec{S} \cdot \vec{I})$ interactions were estimated to be $1.3 \times 10^{-14} \text{ cm}^2$ for Cs-He, and $0.73 \times 10^{-14} \text{ cm}^2$ for Cs-Ne.

6. "Spin Relaxation in Rb-He and Rb-Ne via modification of the Rb hyperfine interaction in van der Waals molecules", F.A. Franz and C. Volk, Phys. Rev. A 18, 599 (1978).

The anomalous modes of electronic spin relaxation previously observed in He and Ne buffer gases were shown to be explained (possibly) by the modification of the Rb hyperfine interaction induced by the formation of Rb-He and Rb-Ne van der Waals molecules. The isolation of anomalous effects was shown also to remove disagreements in cross sections previously reported for the spin relaxation of Cs in Ne.

III. Work partially completed during grant period; submission for publication anticipated in 1979.

1. "Electronic and Nuclear Spin Relaxation in Optically Oriented Potassium Vapor via Sudden and Quasi Molecular Collisions of K Atoms with Noble Gas Atoms", F.A. Franz and C. Volk.
2. "Collisional Relaxation within the 4P States of Potassium Induced by Collisions of K atoms with Noble Gas Atoms", R. Boggy and F.A. Franz.

IV. Persons Participating in this Research**Graduate Research Assistants**

Richard Boggy, thesis, Ph.D., "Studies of Transitions between Zeeman Sublevels of the Potassium 4P States Induced by Non Resonant Thermal Collisions at High Magnetic Fields".

Present employment. Spectra Physics, Mountain View, California.

Charles Volk, thesis, Ph.D., "Investigation of Anomalous Spin Relaxation in Optically Oriented Potassium Vapor".

Present employment. Nuclear Gyroscope Group, Litton Industries

Graduate and Undergraduate Helpers

Ronald Farren, present graduate student, Department of Chemistry, Indiana University.

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